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WASTE ASSAY AND MASS BALANCE FOR THE DECONTAMINATION AND VOLUME REDUCTION SYSTEM AT LANL

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ABSTRACT
The Decontamination and Volume Reduction System (DVRS) operated by the Solid Waste Operations (SWO) Group at Los Alamos National Laboratory (LANL) processes large volume, legacy radioactive waste items. Waste boxes, in sizes varying from 4 ft x 4 ft x 8 ft to 10 ft x 12 ft x 40 ft, are assayed prior to entry into the processing building. Inside the building, the waste items are removed from their container, decontaminated and/or size reduced if necessary, and repackaged for shipment to the Waste Isolation Pilot Plant (WIPP) or on-site low-level waste disposal. The repackaged items and any secondary waste produced (e.g., personal protective equipment) are assayed again at the end of the process and a mass balance is done to determine whether there is any significant hold-up material left in the DVRS building. The DVRS building is currently classed as a radiological facility, with a building limit of 0.52 Ci of Pu239 and Am241, and 0.62 Ci of Pu238, the most common radionuclides processed. This requires tight controls on the flow of nuclear material.

The large volume of the initial waste packages, the (relatively) small amounts of radioactive material in them, and the tight ceiling on the building inventory require accurate field measurements of the nuclear material. This paper describes the radioactive waste measurement techniques, the computer modeling used to determine the amount of nuclear material present in a waste package, the building inventory database, and the DVRS process itself. Future plans include raising the limit on the nuclear material inventory allowed in the building to accommodate higher activity waste packages. All DOE sites performing decontamination and decommissioning of radioactive process equipment face challenges related to waste assay and inventory issues. This paper describes an ongoing operation, incorporating lessons learned over the life of the project to date.

INTRODUCTION
The Decontamination and Volume Reduction System (DVRS) at Los Alamos National Laboratory's (LANL) Solid Waste Operations (SWO) facility was set up to process oversized transuranic (TRU) waste items from Laboratory waste generators. At the start of DVRS operations in 2001, the inventory of existing oversized TRU waste was approximately 2,400 m^3.(1) The oversized waste item packages range in size from 4 ft x 4 ft x 8 ft to 10 ft x 12 ft x 40 ft. Many of these waste containers were stored in earthen bermed storage for up to 20 years and have only recently been retrieved and placed in dome covered storage as part of the Transuranic Waste Inspectable Storage Project.(2) Some common oversize waste containers and their storage configuration are shown in Fig. 1 along with an inset of a box x-ray. The waste items processed in the DVRS include both legacy waste from the Cold War years, and newly generated waste. Most of the waste items are large metal items such as pencil tanks and gloveboxes, but various other items such as large plenum filters are included. The majority of this waste is generated from decontamination and decommissioning activities. The waste is normally packaged either in plywood boxes, fiber reinforced plywood boxes, or metal shipping containers. All of this waste needs to be either

segregated or decontaminated to low-level waste (LLW), or maintained as TRU waste and placed into Waste Isolation Pilot Plant (WIPP) compliant disposal containers. WIPP containers in most cases, means 55-gallon metal drums or WIPP standard waste boxes (SWBs). Much of the waste needs to be size-reduced to fit into the appropriate containers. 75% of the materials processed through the DVRS are expected to be eligible for disposal as LLW, which is far less costly to dispose of than TRU waste. The DVRS building is currently classed as a radiological facility, with a building limit of 0.52 Ci of Pu239 and Am241, and 0.62 Ci of Pu238, the most common radionuclides processed. The limits on the amount of nuclear material which can be inside the DVRS building at any one time require strict radiological inventory controls. Non-destructive assay (NDA) equipment is used along with health physics surveys to keep the building compliant with these limits.

Figure. 1 Common oversize storage containers and example of container x-ray.

THE DVRS PROCESS

The DVRS building is a 13,200 ft^2 confinement structure with active ventilation and contamination control. It includes special containment areas for dismantling the large containers, decontaminating metal objects, and a large shear bailer to volume reduce metal to standard size "pucks" for disposal. The pucks are sized to fit within a 55-gallon drum and can also be placed in an SWB.

The first task for DVRS personnel is to identify the contents (both waste matrix and radiological) of the boxes with existing historical documentation. They then get the boxes x-rayed to verify the contents and get a visual image of the positioning of items in the box, perform head-space gas sampling (for radiological and chemical airborne conditions), and assay the boxes using non-destruction assay (NDA) techniques. After developing a processing plan for the boxes based on the information gathered, DVRS Operations technicians proceed to open the boxes in a confinement area using full level II personal protective equipment (PPE) and standard cutting tools. There is also a HEPA filtered vacuum hose attached to the far side of the box to pull potentially contaminated air away from workers. This air is sampled during processing. Smears and dose rates are taken as needed during processing to determine or verify surface contamination and dose activity levels. These activities are performed under the watchful eyes of industrial hygienists, radiation control technicians (RCTs), and safety personnel.

After opening a box, the (usually plastic wrapped) items are visually examined to determine how the items should be segregated, and whether there are any WIPP prohibited items in the waste or any obvious Resource Conservation and Recovery Act (RCRA) items such as lead or mercury containing items in the containers that need to be classified as mixed waste. The prohibited and RCRA items may be such things as lead bricks, pressurized containers, free liquids, closed containers greater than 4 liters in volume, or lead or mercury contaminated items like light bulbs. These discrete WIPP prohibited or RCRA items are segregated and handled separately. Some items also get individual vents installed in their plastic wrapping to assure compliance with WIPP requirements for hydrogen gas generation rates. Fig. 2 shows an RCT examining waste HEPA filters in a box and an example of a prohibited item being segregated.

Figure 2. Checking a waste for alpha contamination and removal of a prohibited item.

The DVRS processing is proceeding in stages, starting with the least contaminated and best characterized of the waste containers. In Phase 1, 10 containers (31 m^3) were processed. Of those 31 m^3, 27.5 m^3 were segregated and reclassified as LLW. This LLW, consisting of HEPA filters, was sampled and analyzed to show that no RCRA constituents were present and disposed of on site. Phase 2 of operations began in December 2002 and will involve processing more than 250 m^3 of the oversize waste. To date, 60 m^3 of HEPA filters, pencil tanks, metallic debris, and compactable laboratory trash has been processed through the DVRS. Phase 3 will raise the facility limits to that of a nuclear facility (Hazard Category 3), which will allow processing of the remainder of the oversize waste.

RADIOACTIVE ASSAY TECHNIQUES AND MODELING

Waste boxes destined for the DVRS are assayed to determine their radioactive constituents prior to taking the boxes into the facility. After processing, the original waste items and any secondary waste generated during the waste processing are re-assayed using the same technique. This second assay assures a mass balance of nuclear materials entering and leaving the facility and also allows personnel to segregate TRU from LLW items.

The basic NDA technique in the DVRS process utilizes high purity germanium (HPGe) detectors in a far field geometry. A Large Item Neutron Counter (LINC) is utilized for items with adequate amounts of TRU radionuclides to make a large item neutron count of practical use. The HPGe system is very versatile and requires an experienced gamma spectroscopist to set up the measurement parameters. The detector is highly shielded from background radiation and can be placed in the best possible position relative to the waste. The length of the waste measurement varies depending on the container type and the waste matrix. The spectroscopist watches the spectrum develop on a laptop computer and changes the parameters and restarts the measurement if necessary. An example of an HPGe measurement setup for DVRS secondary waste assay is shown in Fig. 3.

Figure 3. HPGe measurement of secondary DVRS waste.

The gamma ray spectra collected during the measurement along with the physical parameters of the measurement are used to determine the radionuclide content of the waste. Usually, the spectra are collected utilizing Ortec®'s Isocart system with Maestro®-32 MCA Emulator, pictured in Fig. 3. The spectra are analyzed using Eberline Services Spectral Nondestructive Assay Platform (SNAP™) analysis code. A review of this characterization methodology is included in the DVRS Phase 1 Technical Defense Report.(4)

INVENTORY CONTROL

The DVRS building is currently classed as a radiological facility with a building limit of 0.52 Ci of Pu239 and Am241, and 0.62 Ci of Pu238. These are the most common radionuclides processed through the DVRS. Other nuclides, generally with lower activity levels, that may be present in the waste items include Pu240, Pu241 (higher activity level, but not TRU), Pu242, Am243, Cs137, Co60, Cm243, Mn56, Np237, Na22, Th228, Th232, U234, U235 and U238. The radiological facility limits on the amount of radioactive material (RAM) inside the DVRS building reflect the conservative DVRS requirement that the cumulative RAM inventory be less than 85% of the Hazard Class 3 (HC-3) threshold limits defined by the DOE.(3) The three phases of the DVRS operations limit the total inventory of RAM as follows:

- Phase 1 limits the RAM to a set of containers whose cumulative inventory is less than 85% of the HC-3 threshold limits,
- Phase 2 limits the RAM to an individual container whose inventory is less than 85% of the HC-3 threshold limits, and
- Phase 3 will involve the operation of the DVRS facility as a HC-3 facility.

In order to determine whether a container or a group of containers plus the facility hold-up and radiological standards/sources meet the building RAM limits, an equation that determines the sum of the fractions of all radiological components is used:

$$\sum_{n=1}^{m} RAM_n / RAM_{nlimit} \leq 0.85 \qquad (1)$$

Where

RAM_n = the measured activity level plus the uncertainty of each radionuclide

RAM_{nlimit} = the HC-3 threshold limit for each radionuclide

m = the total number of radionuclides

This calculation is performed on a near real-time basis by characterizing the waste, including determining the sum of the HC-3 fractions, prior to entry into the DVRS, then characterizing the waste (primary and secondary) upon leaving the facility. Hold-up in the facility is determined based on the inventory difference between the facility input and output values for each container or group of containers. Radiological standards and sources in the building are also counted in the inventory. The four components, the running sum of the input, output (negative), hold-up, and standards/sources together make up the building inventory of RAM. All of this information is tracked on a daily basis using the DVRS Radionuclide Inventory Database.

The isotopic distribution of RAM in plutonium contaminated waste generally falls into one of several categories defined as material types (MTs). Each MT has an average isotopic composition which has been determined based on historical analytical data. In the cases where the RAM characterization process only identifies one or two of the Pu isotopes, the amount of RAM in the remaining isotopes is calculated based on that average distribution. In most cases, the predominant isotope is Pu239, and the remaining alpha-emitting isotopes (Pu238, Pu240, and Pu242) are ratioed based on the Pu239 content. In the cases where only Pu238 is found, that is the only isotope reported, since it represents over 99.9% of the alpha activity in that MT. Where both Pu238 and Pu239 are seen, the remaining isotopes are calculated based on the Pu239 constituent.

At the end of Phase 1, the RAM hold-up calculated from the input and output characterization was determined to be minimal. A comprehensive radiation survey of the building, including the HEPA ventilation units indicated that no detectable activity remained in the building. If there had been any positive smear results, they would have been converted to potential total contamination and added to the information in the Inventory Database. Phase 1 consisted of low activity 4 ft x 4 ft x 8 ft boxes of HEPA filters, with a total HC-3 fraction for all of the boxes of less than 0.85. Fig. 4 shows that for all isotopes the activity of the outgoing material was within the 2-sigma error bounds of the activity of the incoming material.

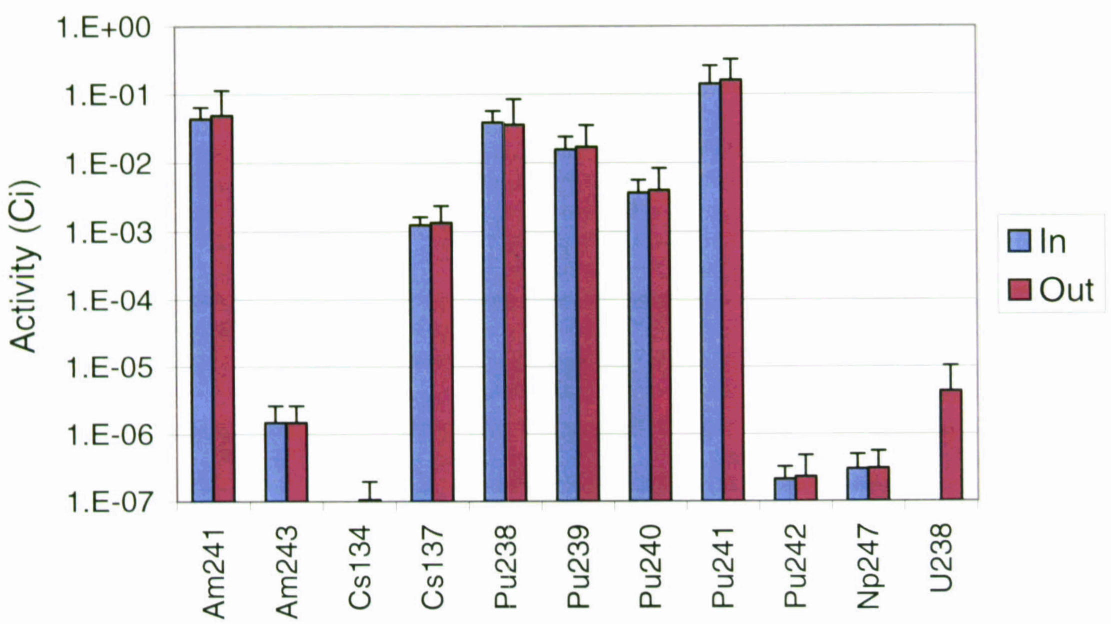

Figure 4. DVRS Phase 1 mass balance.

The database estimates the amount of hold-up in the facility based on the difference between entering and exiting waste. Hold-up is an ever changing number and is created by two processes. The first is a real value caused by the loss of radioactive material from the waste items during processing – onto walls, floors, tools, equipment, PPE, filters, etc. The second is an artificial value created by the inherent differences in the gamma spectroscopy or neutron assay results between the container entering the facility and the primary and secondary waste leaving the facility. This difference arises for several reasons and is accounted for in the assay uncertainty.

One reason for the differences is the differing properties of the entering and exiting containers. The FRP boxes are made up of plywood that has fiberglass secured to the outside of the container with epoxy-like material. These boxes have a somewhat varying wall thickness. Attenuation of gamma rays in the container walls is therefore not entirely consistent from one box to another, or even on different sides of the same box. The primary waste exits the facility in TRU SWB containers, which have metal walls. The secondary waste is generally only wrapped in thick plastic when it is assayed.

An even larger difference can be created by the waste items themselves. Both their density and their position in relation to other items in a container entering or exiting the facility can be factors in the assay uncertainty. Again, attenuation of gamma rays makes a difference in the gamma assay. The position of the RAM contamination in relation to the detector contributes to the uncertainty in the results - that is, is it localized on a side, on top of or on the bottom of an item, are other items between the RAM and the detector, etc. Since the configuration of the waste items changes in processing, the assay results for the entering and exiting materials will be somewhat different. In fact, the exiting material will most certainly have a more accurate assay because the composition and position of the waste items is well-known. Similarly, for neutron assay results, while the location of the RAM is not a factor in many cases, the count rate is affected if there is hydrogenous material in the container (e.g., plastics). Neutron assays of plutonium contaminated material depend on knowing the isotopic composition of the RAM. If this is not well-known, the assay results will be incorrect. Neutron assays are also affected by items which may contain contaminants like plutonium fluoride, or certain non-plutonium radionuclides that spontaneously emit neutrons. In addition, neutron assays are not effective for small amounts of plutonium.

Background radiation can affect the assay results. Background radiation rates vary with the amount of nuclear material (natural or man-made) in the vicinity of the counting equipment, the shielding around the equipment, and in the case of neutron detectors, cosmic ray events. The personnel operating the equipment must remain aware of changing conditions in the background rate and assure that the assay results are not affected.

Finally, all assay calculations are based on counting statistics. Good statistics (i.e., large numbers of counts) produce good results. Longer count times produce better statistics. If count times vary between the entering and exiting waste, the results will vary.

Periodically, the real RAM hold-up in the facility is checked to cancel out these artificial hold-up values. This is accomplished by first removing all known radioactive material from the facility, and adjusting the database for these removals. The pre-filters and roughing filters in the fixed and portable HEPA filter systems and the sawdust generated during container cutting operations are removed and assayed separately. This RAM is also accounted for in the database. Then the facility is radiologically surveyed to determine if actual RAM hold-up exists. If RAM is detected, either the surface will be decontaminated, or the actual hold-up amount will be entered into the database. To be conservative, the database assumes that all gross activity detected is Np237, the radionuclide with the lowest HC-3 threshold value. If no activity is detected in the facility survey, it is assumed that the hold-up indicated by the database is artificial and attributed to assay differences. The RAM inventory is re-aligned to initial conditions and the facility is considered free of hold-up. Three of these surveys have been conducted to date, one at the end of Phase 1 processing, one following the processing of the first six containers in Phase 2, and one after processing the second six containers in Phase 2. All surveys have shown no detectable activity in the building.

CONCLUSION

Tight controls are required to meet the DOE requirements for the various phases of container processing in the DVRS building. The Radionuclide Inventory Database is an effective, near real-time method of controlling the inventory of RAM in the DVRS facility. To date, the three comprehensive RAM surveys have shown that there is no hold-up in the building and that the assay results including uncertainty can be reconciled with the "no hold-up" determination based on the survey results.

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